

Compatibilization of PET-PE blends for the recycling of multilayer packaging foils

Cite as: AIP Conference Proceedings **2055**, 030005 (2019); <https://doi.org/10.1063/1.5084815>
Published Online: 22 January 2019

Laurens Delva, Cédric Deceur, Nicolas Van Damme, and Kim Ragaert



View Online



Export Citation

ARTICLES YOU MAY BE INTERESTED IN

[Recycling of composites - A new approach minimizes downgrading](#)

AIP Conference Proceedings **2055**, 060009 (2019); <https://doi.org/10.1063/1.5084841>

[Characterization of polyamide 6 - polypropylene laminates](#)

AIP Conference Proceedings **2055**, 050008 (2019); <https://doi.org/10.1063/1.5084827>

[Recycled polycarbonate as impact modifier in polypropylene](#)

AIP Conference Proceedings **2055**, 030004 (2019); <https://doi.org/10.1063/1.5084814>

AIP | Conference Proceedings

**Get 30% off all
print proceedings!**

Enter Promotion Code **PDF30** at checkout



Compatibilization of PET-PE Blends for the Recycling of Multilayer Packaging Foils

Laurens Delva^{1,a)}, Cédric Deceur^{1,b)}, Nicolas Van Damme^{1,c)}, Kim Ragaert^{1,d)}

¹*Centre for Polymer and Materials Technologies (CPMT), Department of Materials, Textiles and Chemical Engineering, Faculty of Engineering and Architecture, Ghent University, Technologiepark 915, 9052 Zwijnaarde, Belgium*

^{a)}Corresponding author: Laurens.Delva@ugent.be

^{b)}Cedric.Deceur@ugent.be

^{c)}Ncvdamme.VanDamme@ugent.be

^{d)}Kim.Ragaert@ugent.be

Abstract. The compatibilization of a virgin polyethylene terephthalate/polyethylene (PET/PE) blend and a recycled multilayered post-industrial food tray of similar composition are investigated. The compatibilizers of focus are a propylene ethylene rubber (PER) and a styrene ethylene / butylene styrene triblock copolymer grafted with maleic anhydride (SEBS-g-MA). This work examines the effect of these compatibilizers on both the mechanical properties (impact and tensile properties) and morphology of the virgin blends and verifies whether these effects are transferable to an actual waste stream of post-industrial food trays with comparable composition. Regarding the virgin blends, it was concluded that both compatibilizers increase the impact strength and elongation at break of the material, wherein PER increases the impact strength more effectively than SEBS-g-MA. Moreover, PER succeeds in maintaining both tensile strength and stiffness of the blends. Also, the morphology of the PER blends shows clear indication of rubber toughening. The results of the virgin blends are to a certain extent transferable to an actual waste stream consisting of post-industrial food trays with comparable composition. PER shows to be the most promising candidate for increasing toughness while maintaining stiffness and tensile strength.

INTRODUCTION

In multilayered packaging materials, different polymers are stacked in layers in one foil or sheet to maximize the performance of the packaging material. Each polymer contributes in his own way to the overall technical functionality. Polyethylene terephthalate (PET) is often used for water and gas barrier and mechanical strength, while polyethylene (PE) is often used because of its excellent sealing properties, water barrier properties and low-temperature performance. Common combinations in multilayered foils and sheets are PET/PE, polyamide (PA)/PE and PET/PE/ethylene vinyl alcohol (EVOH)/PE¹.

Since the different polymers are physically connected, it is very difficult or economically not feasible to separate them from another. This implies they will have to be mechanically recycled together. Due to the low miscibility of these polymers, the blends have in turn inferior mechanical properties. According to Plastics Recyclers Europe annually 700000 tonnes of PET containing food trays are not sorted out and not effectively recycled in Europe². In order to be able to use this waste stream for new closed-loop or open-loop products, the mechanical properties must be improved. This can be done either by using additives that improve miscibility or specific custom processing techniques³.

Mainly, we can divide the multilayered packaging waste in post-industrial and post-consumer waste. Post-industrial waste typically is production waste coming from foil producers (e.g. cutting edges, changeovers or non-conform products) and packagers (e.g. unfilled trays, leftovers or cutting edges). This waste is typically quite clean and uncontaminated. Post-consumer waste, however, is mostly still contaminated with organics or other plastics⁴.

The purpose of this study is to examine whether the mechanical properties of a recycled post-industrial waste stream consisting of PET trays and films can be improved by adding propylene ethylene rubber (PER) or styrene ethylene / butylene styrene grafted with maleic anhydride (SEBS-g-MA). The influence of these additives on the morphology is also investigated.

Firstly, virgin components are used to simulate the selected waste stream. The virgin blend consists of PET and PE and is processed with and without the compatibilizers. These blends are then mechanically characterized with tensile and impact tests. Furthermore, the morphology is determined using Scanning Electron Microscopy (SEM). Finally, a real waste stream is used consisting of post-industrial food trays. This waste stream is also processed with and without compatibilizers and then mechanically characterized. As with the virgin blends, morphology is also determined using SEM. It is intended to determine the effect of the compatibilizers on an actual waste stream as well as the transferability of the results of the virgin blends to this waste stream.

EXPERIMENTAL

Materials

The selected PET used as matrix is Lighter™ C93 produced by Equipolymers. This PET is made of purified terephthalic acid (PTA) and monoethylene glycol (MEG). It has a melting temperature of 247 °C and a glass transition temperature of 78°C. For PE, Exceed™ 1012 HA mVLDPE produced by Exxonmobil is used. It consists of a metallocene ethylene-hexene copolymer and is designed for the production of sheets. The used PE has an MFI of 1.0 g/10 min and melting temperature of 115°C. The used SEBS-g-MA as compatibilizer is FG1901 G Polymer produced by Kraton. The grafting level is 1.4 to 2.0 wt%. The used PER is Vistamaxx™ 6202 produced by Exxonmobil. It is primarily composed of isotactic propylene repeat units (85 wt%) with random ethylene distribution (15 wt%). Postindustrial food trays were offered by Belgian packaging company Ter Beke and consist predominantly of PET and PE.

Sample Preparation

Samples were prepared by melt blending, followed by injection moulding. The blend components were dried at 60°C during 6h in a vacuum dryer prior to melt processing. The dried pellets were mixed in the weight ratios shown in Table 1. The food trays were shredded before they were fed into the extruder for melt blending. The melt blending of the different mixtures was performed on a co-rotating twin-screw extruder Coperion ZSK 18 MEGAlab. The temperature profile starting from the feeding zone to the die was 190°C – 240°C – 245°C – 245°C – 250°C – 250°C – 255°C – 260°C. The screw speed was maintained at 200 rpm. The extrudate was cooled down and stranded in a water bath before pelletization. Before injection moulding, the pellets were dried again at 60°C for 6 hours.

Samples for impact (ISO 179 type 1) and tensile (ISO 527 type 1A) testing were injection moulded with an Engel ES 330 / 80 HL with barrel temperatures spanning the range 250°C to 280°C and the mould temperature at around 15-20°C.

TABLE 1. Composition of the samples.

		PET [wt%]	PE [wt%]	rTray [wt%]	SEBS-g-MA [wt]	PER [wt%]
Virgin	PET/PE	80	20	-	-	-
	PET/PE/SEBS-g-MA-2.5	78	19.5	-	2.5	-
	PET/PE/SEBS-g-MA-5	76	19	-	5	-
	PET/PE/PER-5	76	19	-	-	5
Recycled	rTray	-	-	100	-	-
	rTray/SEBS-g-MA-5	-	-	95	5	-
	Tray/PER-5	-	-	95	-	5

Characterization

Scanning electron microscopy (SEM) was used to study the surface morphology of the samples using a JSM7600F FEG SEM. The brittle fracture surfaces of the samples were examined. The brittle fracture was achieved by cryogenic breaking after liquid nitrogen cooling. The samples were sputtered with gold by a Baltec SCD005 sputter coater. The accelerating voltage was 15-20 kV.

Charpy impact properties (ISO 179 notched) were measured using a Tinius Olsen IT 503 Pendulum Impact Tester, equipped with a hammer with an energy of 2 J.

Tensile properties (ISO 527) were determined using an Instron 5565 tensile testing machine, with a load cell of 5 kN. The dynamometer was equipped with a Dynamic Extensometer 2620-603 by Instron, which was used to accurately measure the strain. The crosshead speed was set at 1 mm/min to determine the tensile properties until yielding. Then the crosshead speed was increased to 25 mm/min until failure.

All mechanical tests were performed at 23°C and 50% relative air humidity. All the reported results are the mean of at least ten measurements. Statistical analysis of all results was performed by the software package SPSS Statistics 22 (t-independent sample tests, $p=0.05$).

RESULTS

Mechanical properties

Table 2 gives an overview of the mechanical properties of the different virgin and recycled blends. The impact test shows that the addition of both SEBS-g-MA and PER has a positive influence on the toughness of the virgin blends indicated by an increase in strain at break (ϵ_b) and impact strength. The addition of 5 wt% SEBS-g-MA and PER respectively doubles and triples the value of the impact strength in comparison with the virgin PET/PE blend. In case of the SEBS-g-MA, both the tensile modulus (E_t) and the tensile stress (σ_t) are significantly reduced upon the addition of the compatibilizer. This reduction indicates a poor effectiveness of the compatibilizer. PER, on the other hand, succeeds in maintaining both tensile strength and stiffness in comparison with the virgin blend. A plot of stiffness and impact strength for the virgin blends is shown in Figure 1a. These results propose PER as the ideal candidate to increase toughness while maintaining stiffness and tensile strength for this specific blend.

The results of the mechanical tests of the virgin blends are to a certain extent transferable to the recycled post-industrial trays. It is seen that PER similarly increases the strain at break and impact resistance but the increase is less pronounced. Adding SEBS-g-MA, however, does not cause a significant increase in impact resistance. The strain at break in the rTray/SEBS-g-MA-5 blend reaches yet the highest value, which indicates there is some degree of compatibility. The results show that the recycled blends perform better than their virgin equivalents despite their comparable composition. A possible explanation is the usage of a different grade of PET and PE which affects compatibility between PET and PE and changes the starting mechanical properties of the materials⁵. A plot of stiffness versus impact strength for the recycled blends is shown in Figure 1b. PER remains the preferred candidate to increase the toughness of the recycled trays despite a slight decrease in stiffness.

TABLE 2. Mechanical properties of the blends with indication of the standard deviation.

		Notched impact [kJ/m²]	E_t [MPa]	σ_t [MPa]	ϵ_b [%]
Virgin	PE	> 100	95 ± 9	15.22 ± 0.49	571.46 ± 13.10
	PET	2.17 ± 0.19	2479 ± 129	41.01 ± 6.01	2.47 ± 0.72
	PET/PE	2.15 ± 0.07	1409 ± 36	28.07 ± 0.40	4.55 ± 1.19
	PET/PE/SEBS-g-MA-2.5	3.33 ± 0.29	1307 ± 58	26.64 ± 0.54	8.62 ± 2.37
	PET/PE/SEBS-g-MA-5	4.14 ± 0.43	1280 ± 57	25.75 ± 0.48	34.94 ± 18.05
	PET/PE/PER-5	6.24 ± 0.16	1409 ± 37	27.41 ± 0.54	41.54 ± 15.90
Recycled	rTray	3.99 ± 0.32	1677 ± 91	31.40 ± 1.24	14.14 ± 10.20
	rTray/SEBS-g-MA-5	4.63 ± 0.56	1550 ± 50	31.97 ± 0.75	355.57 ± 24.73
	Tray/PER-5	5.78 ± 0.20	1532 ± 80	28.35 ± 0.85	23.58 ± 6.91

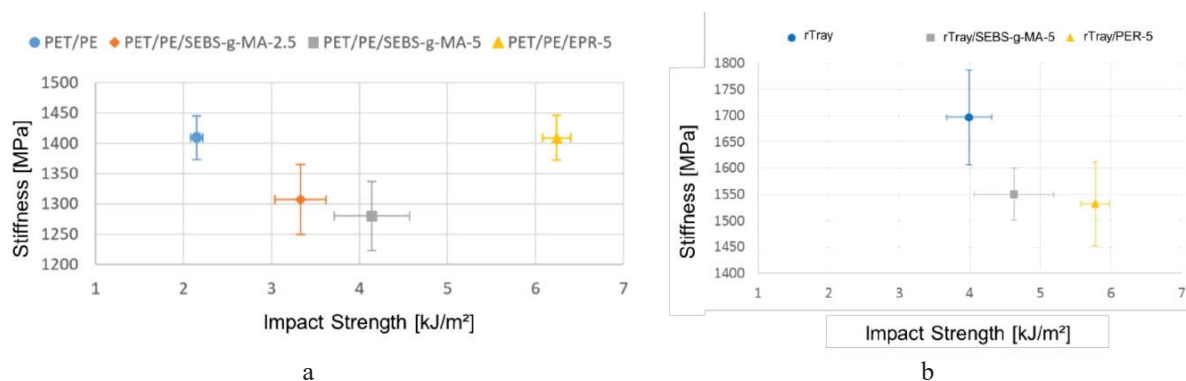


FIGURE 1. Plot of stiffness versus impact strength for (a) virgin and (b) recycled tray blends.

Morphology

Figure 2 shows the SEM-micrographs of the different blends. Figure 2a displays the morphology of a typical immiscible blend in which PE droplets are dispersed in the PET matrix. In the virgin blends (Figures 2a, b and c) there is no clear improvement in morphology by the addition of compatibilizer. Most results from literature focus on the polar component being the minor phase, while in this research it is used as matrix polymer. The average size of the dispersed PE particles remains the same. Both additives show no compatibilization effect at the interface. In case of the PER blend, the presence of a new, fine morphology in the PET matrix can be noticed. This finely divided rubber phase with small interparticle distances can explain why PER performs better in terms of impact strength than SEBS-g-MA⁶.

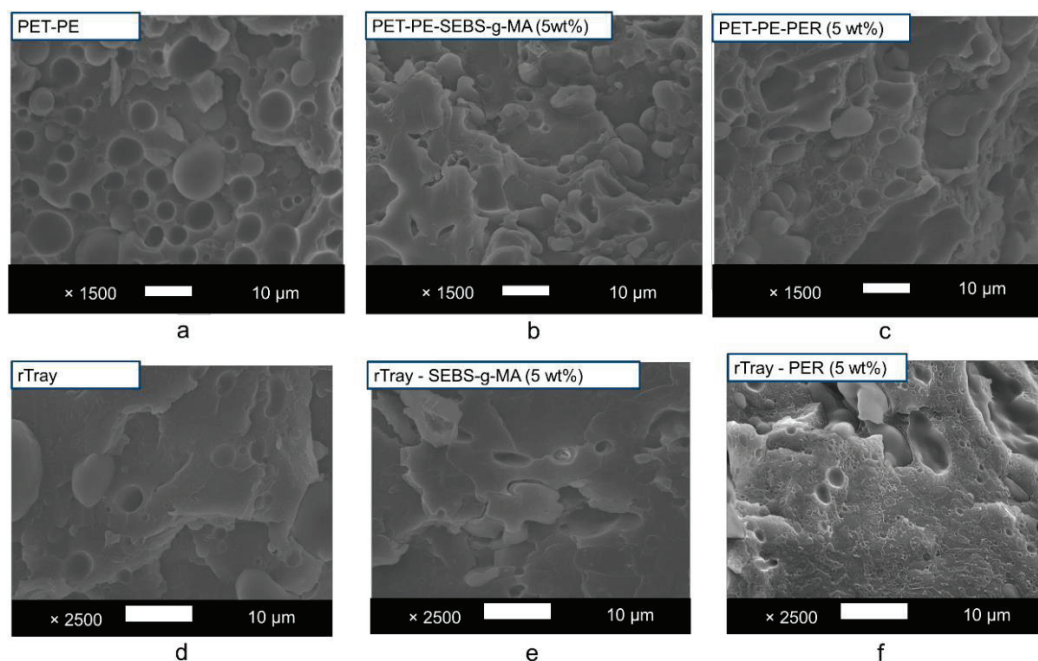


FIGURE 2. SEM-micrographs of the different blends: (a) PET/PE, (b) PET/PE/SEBS-g-MA-5, (c) PET/PE/PER-5, (d) rTray, (e) rTray/SEBS-g-MA-5 and (f) rTray/PER-5

In case of the compatibilized recycled blends (Figures 2e and f), the size of the dispersion particles appears to be as large as the recycled trays (Figure 2d), confirmed by determining the particle size with imaging software. This indicates that in the recycled blends the compatibilizers similarly fail to reduce the size of the dispersed phase. Once

again the presence of a very fine dispersion phase can be seen at the recycled blend to which PER was added. Consequently, it is plausible to assume that both PER and SEBS-g-MA have an impact-modifying rather than a compatibilizing effect, confirmed by the mechanical results.

CONCLUSIONS

This work examines the effect of two compatibilizers SEBS-g-MA and PER on the mechanical properties (impact and tensile properties) and morphology of an 80/20 PET-PE virgin blend and verifies whether these effects are transferable to an actual waste stream of post-industrial food trays with comparable composition. Regarding the virgin blends, it can be concluded that both compatibilizers increase the impact strength and elongation at break of the material, wherein PER increases the impact strength more effectively than SEBS-g-MA. Moreover, PER succeeds in maintaining both tensile strength and stiffness of the blends. Besides the mechanical performance, the morphology of the PER blends shows clear behavior of rubber toughening. The results of the virgin blends are to a certain extent transferable to an actual waste stream consisting of post-industrial food trays with comparable composition. PER also increases the toughness of the recycled food trays. However, the increase is less significant and a small decrease in stiffness can be noticed. Overall, PER shows to be the most promising candidate for increasing toughness while maintaining stiffness and tensile strength. To conclude, these results look promising for the recycling of multilayered plastic waste streams.

REFERENCES

1. J. R. Wagner Jr, *Multilayer flexible packaging*. (William Andrew, 2016).
2. E. Tarlowska, *PET trays existential dilemma*. (Plastics Recyclers Europe, 2014).
3. C. Koning, M. van Duin, C. Pagnoulle and R. Jerome, *Prog. Polym. Sci.* **23** (4), 707-757 (1998).
4. S. Huysman, J. De Schaepmeester, K. Ragaert, J. Dewulf and S. De Meester, *Resources, Conservation and Recycling* **120**, 46-54 (2017).
5. Y. Zhang, H. Zhang, W. Guo and C. Wu, *Polymers for advanced technologies* **22** (12), 1851-1858 (2011).
6. G. Sarkhel, A. B. and A. Banerjee, *Polym. Plast. Technol. Eng.* **49** (10), 1049-1054 (2010).